

Recovery of surfactant SDS and Cd²⁺ from permeate in MEUF using a continuous foam fractionator

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Received 26 June 2007; received in revised form 11 November 2007; accepted 12 November 2007

Available online 17 November 2007

Abstract

The purpose of this study was to investigate the use of foam fractionation to recover valuable surfactant (SDS) and metal ion (Cd²⁺) in the permeate of micellar-enhanced ultrafiltration (MEUF). The effects of operating factors, such as time, air flow rate, feed flow rate, liquid height, foam height, feed surfactant concentration, ethanol concentration and temperature on separation characteristics were studied in the continuous operation. When the concentrations of surfactant (SDS) and metal ion (Cd²⁺) in the feed solution were 500 mg/L and 10 mg/L, an enrichment ratio of 3.1 was achieved for SDS along with 52% removal fraction, as well as 99.35% Cd²⁺ was removed, after optimization of different process parameters. As the optimized results, the air and liquid flow rates were 100 L/h and 5 L/h, liquid and foam heights were 45 cm and 66 cm, respectively, sparger pore size was 10 μm. The Cd²⁺ concentration in the effluent was lower than 0.1 mg/L which could meet the integrated wastewater discharge standard (the first grade of national discharge standards in China).

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Keywords: Foam fractionation; Surfactant (SDS); Metal ions (Cd²⁺)

1. Introduction

Surfactant-based process such as micellar-enhanced ultrafiltration (MEUF) has been successfully used to remove metal ions and/or soluble organic solutes from aqueous streams [1–4]. When a surfactant is added into polluted aqueous, it forms micelles at a concentration higher than its critical micellar concentration (CMC). Metal ions bind to the surface of negatively charged micelles of an anionic surfactant while organic solutes tend to dissolve or to be solubilized within the micelles. The mixture is then forced through an ultrafiltration membrane with pore sizes small enough to block passage of the micelles associated with metal ions and/or dissolved organics. However, the surfactant monomers which did not form micellars and were not attracted by metal ions or unsolubilized organic molecules passed through the membrane. Therefore, the permeate contains small amount of surfactant, metal ions and organics.

Since the MEUF was proposed, there were lots of studies about its application in the wastewater treatment. However, these studies mostly focused on how to obtain high rejection for the removal of dissolved contaminants by MEUF. Few studies have been conducted on the leakage of surfactant monomers (surfactant molecules not in the micelle form) through the ultrafiltration membrane. This can add substantial expense to the separation or make the process effluent stream environmentally unacceptable [5–7].

Foam fractionation is a simple and low-cost method, belonging to the adsorptive bubble separation techniques [8]. In foam fractionation, the more surface-active compounds are attached by adsorption to gas bubbles, which then rise to the top of the liquid at the surface, the less surface-active molecules remained entrapped in the interstitial liquid (Fig. 1). When these bubbles rise out of the solution, foam is formed. Only a small fraction of liquid is carried with the bubbles into the foam phase due to gravity drainage. The foam phase can be collapsed into a new liquid foamate by releasing the gas bubbles. The concentration of the surface-active solute in the new liquid is usually several times of that in the initial liquid solution.

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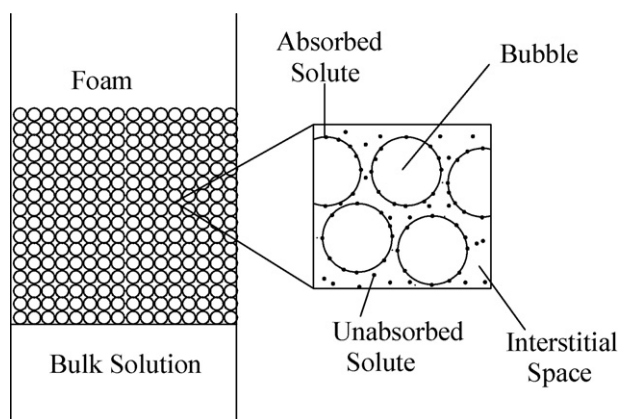


Fig. 1. Phenomena occurring during foam fractionation.

Foam fractionation is rapidly becoming an effective method to separate surface-active and non-surface-active species. There have been many studies on foam fractionation including the effect of operating mode [9,10] and foam properties [11,12]. Foam fractionation has been extensively studied for the purpose of removing pollutants such as heavy metal ions and proteins from water by adding surfactant [13–16]. Rubin et al. [17] investigated the removal of lead (II) using sodium lauryl sulfate as a collector. Several experiments have also been done to investigate the recovery of surfactant itself using foam fractionation. Boonyasuwat et al. [18] studied the efficiency of foam fractionation for cationic surfactant cetylpyridinium chloride (CPC) removal from water. Many variables were considered to have a significant effect upon removal efficiency, such as the air flow rate, the bubble diameter, feed concentration, foam height and temperature. Despite this large number of publications, the use of foam fractionation for recovery of valuable surfactant and metal ions in permeates of MEUF has seldom been reported.

In this study, we designed and built a single continuous foam fractionator and investigated a continuous operation in the recovery of SDS and Cd²⁺ from the permeate in MEUF. The effects of conditions of feed solution (time, pH, ethanol concentration, temperature) and the operational parameters of the column (air and feed flow rate, liquid and foam phase height) on the separation characteristics were investigated.

2. Materials and methods

2.1. Materials

Anionic surfactant, sodium dodecyl sulfate (SDS) with a purity of 99% was obtained from Sigma Chemical Reagents Co., Ltd. (Shanghai, China); Ethanol was AR Grade purchased from BDH Chemicals Ltd. (Poole, England). All of the above materials were used without further purification. Distilled water was used in all experiments.

2.2. Equipment

Fig. 2 is a schematic of a simple continuous mode foam fractionation device. The column consisted of a cylindrical glass

tube (14 cm inside and 190 cm in length) with an inlet at the top and one outlet underneath, and the thickness of 2 mm. Four foam heights of 22 cm, 44 cm, 66 cm and 88 cm from the top of the column were studied. Five liquid sample ports were located at the different positions of cylindrical glass tube. The feed stream tube, drainage stream tube, liquid sample and foam outlet tube were made of acrylic with 1 cm o.d.

2.3. Methods

At the beginning of each run, the feed solution was placed in the column at a height of 12 cm. Then the pressurized air was sparged into the column through sintered glass diffuser with pore size of 10 μm at the bottom of the column. The feed rate was pumped through a flowmeter at 5 L/h by a metering pump and the liquid height in the column was controlled by adjusting the flow of outlet of a tube leading from the lower end of the column. Afterwards, additional solution was fed into the column from the feed storage tank continuously. The feed temperature was kept between 20 °C and 25 °C in all runs. Foam from the column top and liquid in different positions was collected at different heights after a designed time interval. The foam was frozen to prevent loss of water and thawed to obtain the collapsed foamate samples.

The foam fractionation was studied under steady-state. The base conditions were as follows: 5 L/h liquid feed rate, 100 L/h air flow rate, 66 cm foam height, and 45 cm liquid height. When the initial concentrations of SDS and Cd²⁺ applied in MEUF were 900 mg/L and 50 mg/L, their concentrations in the permeate were 500 mg/L and 10 mg/L, respectively [19]. All runs were carried out for a minimum of 3.5 h. Steady-state was ensured when all measured parameters were invariant with time. During the experiment, the surfactant concentration (mg/L) and metal ion (Cd²⁺) concentration in the collapsed foam solution were measured. The CMC of SDS (7.8 mmol/L) was obtained by conductivity measurement (not shown).

2.4. Analytical methods

The concentration of SDS was measured by the methylene blue spectrophotometric method with Shimadzu UV-2550(P/N206-55501-93) spectrophotometer from Japan. The concentration of cadmium ions was analyzed by atomic adsorption spectroscopy (Perkin-Elmer, Modle AAnalyst 700).

2.5. Calculations

The performance of foam fractionation is commonly expressed in two terms: enrichment ratio E ; and removal fraction R .

$$R(\%) = \frac{C_i - C_e}{C_i} \times 100 \quad (1)$$

$$E(\%) = \frac{C_f}{C_i} \times 100 \quad (2)$$

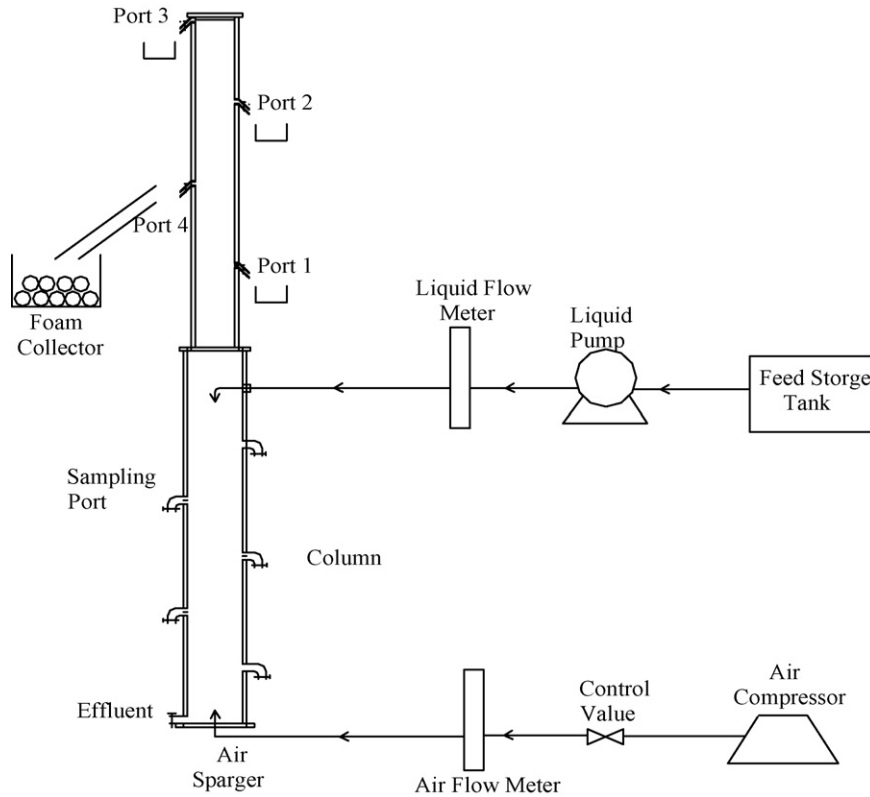


Fig. 2. Schematic diagram of the continuous foam fractionation system.

where C_i and C_e are the surfactant or metal ion concentrations (mg/L) in the influent and effluent streams, respectively, and C_f is the surfactant or metal ion concentration in the collapsed foam solution (mg/L).

3. Results and discussion

3.1. Effect of time

The effect of the time on the separation characteristics was investigated (Fig. 3). The enrichment ratio decreased while removal fraction increased with time. Stable operation is attained after 210 min, where the removal fraction of SDS and Cd^{2+} converge to 52% and 99.35%. Steady-state was attained after 210 min and ensured when all measured parameters were invariant with time.

3.2. Effect of air flow rate

The effect of the air flow rate on the separation characteristics was investigated (Fig. 4). Along with the air flow rate increased, enrichment ratio decreased, while removal fraction increased. These observations agree with those of Brown et al. [20]. The air flow rate was adjusted stepwise from 60 L/h to 240 L/h, SDS enrichment ratio decreased from 5.1 to 2, and removal fraction went up from 26% (at 60 L/h) to 52% (at 100 L/h), then decreased again to 32% at 240 L/h (Fig. 4a). With the increase of air flow rate from 60 L/h to 240 L/h, the Cd^{2+} enrichment

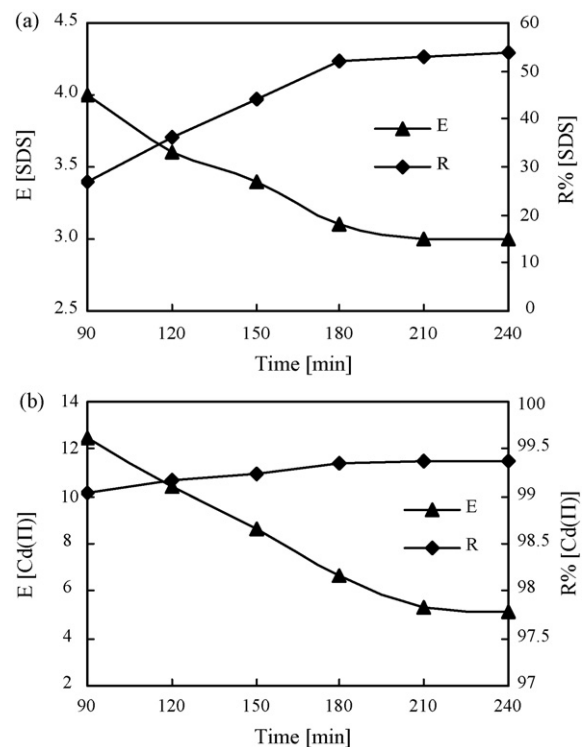


Fig. 3. Effect of time on surfactant separation efficiency (a) and Cd^{2+} separation efficiency (b). Initial concentrations of SDS and Cd^{2+} were 500 mg/L and 10 mg/L, respectively; foam height, 66 cm; liquid height, 45 cm; liquid flow rate, 5 L/h; air flow rate, 100 L/h.

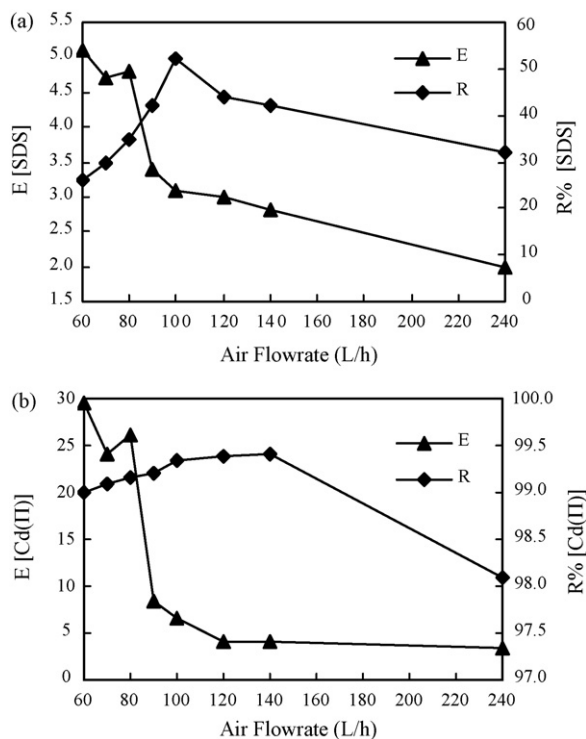


Fig. 4. Effect of air flow rate on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of SDS and Cd²⁺ were 500 and 10 mg/L, respectively; foam height, 66 cm; liquid height, 45 cm; liquid flow rate, 5 L/h.

ratio decreased from 29.5 to 3.5, while the removal fraction increased to a maximum value of 99.42%, then decreased again to 98.1% at 240 L/h (Fig. 4b). It can be explained as follows: with the rise of air flow rates, more bulk liquid could be transported into the foam and adsorbed onto the bubble surfaces, so the bubble production increased. But the lower residual time of bubbles in foam phase causes lower drainage of liquid from foam, so there is a higher content of water in foamate. Also at high air flow rate, bubbles move up rapidly to the top of the column because of the higher liquid entrainment rate. This effect was more dominant than that of the decrease in foam concentration, so the removal fraction increased with the air flow rate.

3.3. Effect of foam height

Results of the foam height study can be found in Fig. 5. As foam height increased, SDS enrichment ratio increased and removal fraction decreased (Fig. 5a); while Cd²⁺ enrichment ratio increased and removal fraction remained constant (Fig. 5b). As the foam height increased, a longer foam residence time which allowed for more drainage of the liquid in the films, resulted in a dryer foam and higher enrichment ratio. With the increase of foam height, the surfactant removal fraction decreased, because of the increased rate of foam collapse resulted from foam drainage. Similar results have previously been published [21]. However, in the range of foam heights from 22 cm to 88 cm, the relative constant removal fraction was higher

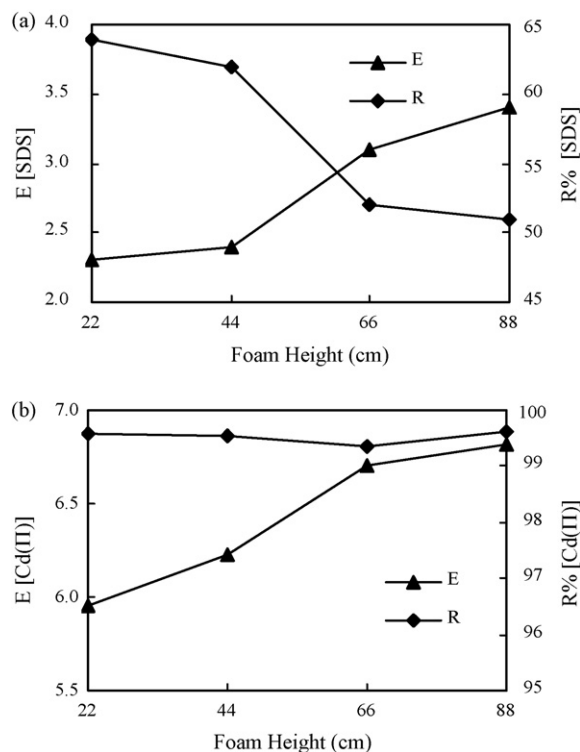


Fig. 5. Effect of foam height on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of SDS and Cd²⁺ were 500 mg/L and 10 mg/L, respectively; air flow rate, 100 L/h; liquid height, 45 cm; liquid flow rate, 5 L/h.

than 99% and no significant effect on Cd²⁺ removal fraction was found.

3.4. Effect of liquid height

Fig. 6 shows the effect of the liquid height on the removal fraction and the enrichment ratio of SDS and Cd²⁺ under steady-state condition. As the liquid height increased, the surfactant and Cd²⁺ enrichment ratio both went up, while the removal fraction remained constant around unity. The results indicated that increasing liquid height led to the increase of residence time of bubbles in the liquid. It took longer for bubbles to rise through the solution, and adsorption of surfactant at the gas–liquid interface could approach an equilibrium level more closely, and raise the enrichment ratio. However, liquid height had little effect on removal fraction over the range studied, and the removal fraction of Cd²⁺ was higher than 99% (Fig. 6b).

3.5. Effect of feed flow rate

The effect of the feed flow rate is shown in Fig. 7. An increase in the feed flow rate resulted in decreasing the enrichment ratio. A similar trend between feed flow rate and enrichment ratio was also observed in the previous work [22,23]. The feed flow rate was adjusted from 3 L/h to 10 L/h, SDS enrichment ratio decreased from 3.3 to 2.5, and the removal fraction decreased from 52% to 30% (Fig. 7a). It can be explained as follows: at higher feed flow rates the lower residual time of bubbles in

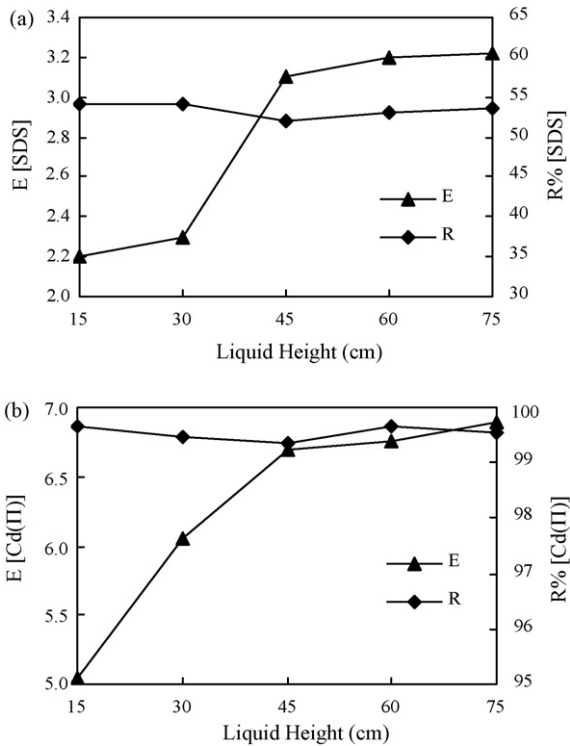


Fig. 6. Effect of liquid height on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of SDS and Cd²⁺ were 500 mg/L and 10 mg/L, respectively; air flow rate, 100 L/h; foam height, 66 cm; liquid flow rate, 5 L/h.

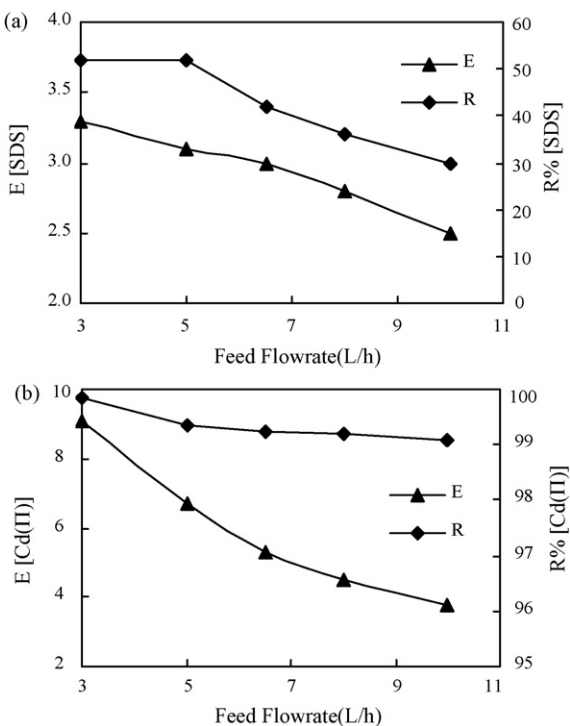


Fig. 7. Effect of feed flow rate on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of SDS and Cd²⁺ were 500 mg/L and 10 mg/L, respectively; air flow rate, 100 L/h; foam height, 66 cm; liquid height, 45 cm.

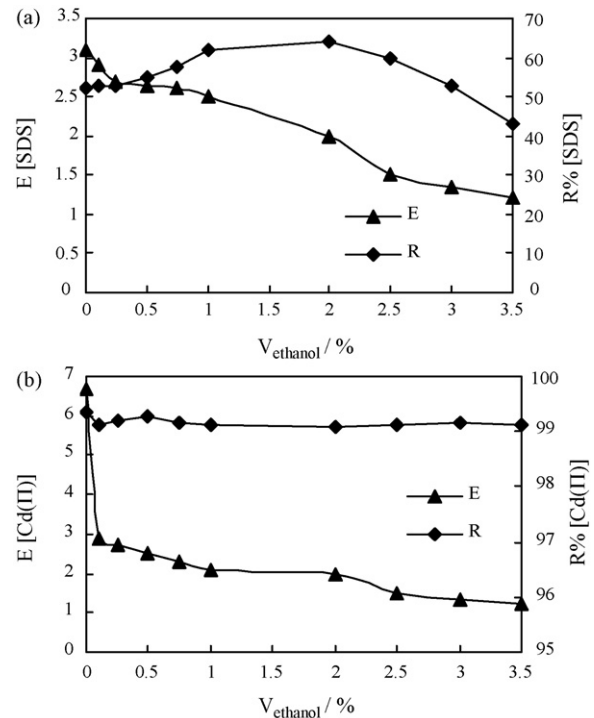


Fig. 8. Effect of ethanol amount on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of SDS and Cd²⁺ were 500 mg/L and 10 mg/L, respectively; air flow rate, 100 L/h; liquid height, 45 cm; foam height, 66 cm; liquid flow rate, 5 L/h.

the column causes lower drainage, decreasing the enrichment ratio. Fig. 7b shows that with the feed flow rate increasing, the Cd²⁺ enrichment ratio was reduced from 9.1 to 3.8. The removal fraction of Cd²⁺ was higher than 99%, which means feed flow rate had little effect on it.

3.6. Effect of ethanol concentration

It is well known that the addition of small amounts of ethanol in water leads to inhibition of the coalescence tendency of small bubble which has the effect of stabilizing the homogeneous bubble flow regime [24,25]. Study results of the ethanol concentration could be found in Fig. 8. As the concentration of ethanol increased, the SDS enrichment ratio decreased from 3.1 to 1.2, while the removal fraction went up from 52% to a maximum value of 64%, then decreased again to 43% (Fig. 8a). Fig. 8b shows that with the increasing concentration of ethanol, the Cd²⁺ enrichment ratio was reduced from 6.7 to 1.25, while removal fraction approached unity. This may be due to the reduction in the surface tension of the thin liquid film lamellae and the increase in solution viscosity with the concentration of ethanol increasing, which can form some stable and small bubbles. The enhanced stability of foam with small bubble was correlated with the decrease on drainage.

The increasing ethanol concentration resulted in the increase of surfactant removal fraction because of more adsorption of solution onto the bubble surfaces as small bubbles slowly rose up to the top of column. While with a further increase in ethanol concentration, the surfactant removal fraction decreased again.

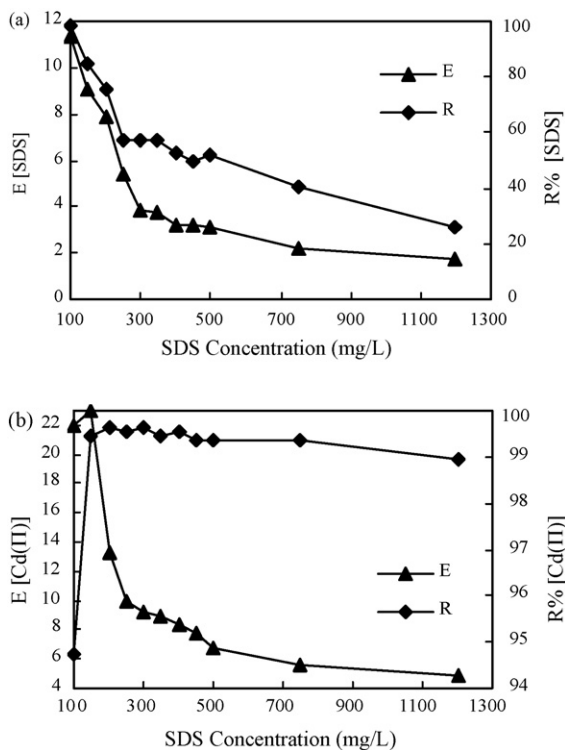


Fig. 9. Effect of SDS concentration on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of Cd²⁺ was 10 mg/L; air flow rate, 100 L/h; liquid height, 45 cm; foam height, 66 cm; liquid flow rate, 5 L/h.

This was perhaps because less adsorption onto the bubble surface happened since ethanol occupied too much interface. The concentration of ethanol had little effect on Cd²⁺ removal fraction over the range studied.

3.7. Effect of surfactant concentration

The concentration of surfactant (SDS) in the feed solution has also been found to significantly influence the performance of foam fractionation. As seen from Fig. 9a, the increasing concentration of the surfactant resulted in the reduction in the enrichment ratio as well as removal fraction of SDS. It was caused by lower surface tension and higher surface liquid viscosity which caused the rate of film drainage decreased. The contribution of adsorbed surfactant to enrichment ratio decreased since the amount of surfactant in the bulk liquid was larger. The increasing concentration of surfactant resulted in a lower removal fraction of SDS, presumably due to the saturation of the adsorbed surfactant at the air–water interface. As shown from the experimental consequence, to realize high enrichment ratio and removal fraction values, foam fractionation should be used at lower surfactant concentrations.

Fig. 9b also shows that with an increase of surfactant concentration from 100 mg/L to 1200 mg/L, the Cd²⁺ enrichment ratio was reduced from 22.05 to 4.85, while the removal fraction increased up to a maximum value of 99.62%, then decreased again to 98.94% (at 1200 mg/L). The former was caused by higher foam stability and lower surface tension at increasing

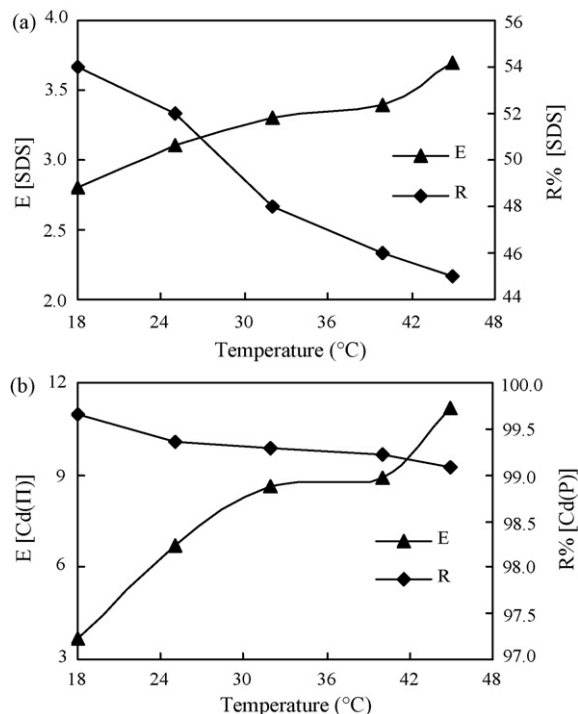


Fig. 10. Effect of temperature on surfactant separation efficiency (a) and Cd²⁺ separation efficiency (b). Initial concentrations of SDS and Cd²⁺ were 500 mg/L and 10 mg/L, respectively; air flow rate, 100 L/h; liquid height, 45 cm; foam height, 66 cm; liquid flow rate, 5 L/h.

surfactant concentration in the foam. As a consequence of the lower surface tension, liquid-keeping was increased. However, the concentration of the surfactant cannot be infinitely enhanced. Once the surface is saturated, further increase in surfactant concentration will significantly slow down the rate of drainage of the foam, and surplus surfactant can form the micelles. Therefore, the Cd²⁺ removal fraction reduced again at the surfactant concentration of 1200 mg/L.

3.8. Effect of temperature

Fig. 10 shows the effect of temperature on the separation characteristics. Increasing the temperature, the enrichment ratio increases, perhaps because drainage rates increase as viscosity decreases, resulting in decreasing water content in the thin liquid film. With the increase of the temperature, the rate of the surfactant removal fraction decreased. It is maybe that the decrease rate in the collapsed foam flow is much higher than the increase rate in the surfactant concentration as the temperature increases. However the temperature had little effect on the Cd²⁺ removal fraction, the Cd²⁺ removal fraction (99%) was obtained.

4. Conclusions

Foam fractionation is effective in the recovery of valuable surfactant (SDS) and micro metal ion (Cd²⁺) in permeates of MEUF process. The parameters include air and feed flow rate, liquid

and foam heights, ethanol concentration, time, influent surfactant concentration and temperature which were investigated in single-stage continuous operation. When the concentrations of SDS and Cd^{2+} in the feed solution were 500 mg/L and 10 mg/L, the air and liquid flow rates were 100 L/h and 5 L/h, liquid and foam heights were 45 cm and 66 cm, respectively, sparger pore size was 10 μm , and then an enrichment ratio of 3.1 was achieved for SDS along with 52% removal fraction, as well as the enrichment ratio and removal fraction of Cd^{2+} was 6.7 and 99.35%, respectively. The concentration of Cd^{2+} was lower than 0.1 mg/L in the effluent, which could meet the integrated wastewater discharge standard. Changes in air flow rate had a strong effect on the enrichment ratio and removal fraction of the SDS and Cd^{2+} . An increase in the feed flow rate, ethanol and surfactant concentration resulted in a decrease in the enrichment ratio for both SDS and Cd^{2+} . Therefore, raising these parameters has a negative impact on foam fractionation as less concentrated foam liquid is recovered overhead. Liquid height, foam height, feed flow rate, ethanol concentration and temperature have little effect on Cd^{2+} removal fraction over the range studied, and the removal fraction of Cd^{2+} was higher than 99%.

Acknowledgements

This study was financially supported by the National 863 High Technology Research Program of China (2004AA649370), the National Basic Research Program (973 Program) (2005CB724203), the National Foundation for Distinguished Young Scholars (50225926, 50425927), the Doctoral Foundation of Ministry of Education of China (20020532017) and the Teaching and Research Award Program for Outstanding Youth Teachers in Higher Education Institutions of MOE, P.R.C. (TRAPOYT) in 2000, the National Science Foundation of China (No. 50608028), the Scientific Research Fund of Hunan Provincial Education Department ([2005]100).

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